

REMARKS

Claims 1-4 and 10 currently are pending. Claim 1 currently has been amended. Claim 10 newly has been added. Claims 5-9 have been canceled.

Claims 1-9 are rejected under the judicially created doctrine of obviousness-type double patenting as being unpatentable over claims 1-16 of US 6,649,709.

To overcome this rejection applicants herein file a terminal disclaimer.

Claims 1-4 are rejected under 35 USC § 103(a) as being unpatentable over Mueller et al. (US 5,324,697), or Smith et al. (US 4,587,227), each individually in view of Hennenberger et al. (EP 0571 826). The examiner believes one of ordinary skill in the art would be motivated by the expectation of success to develop a similar set of equation to have a better control on the densities of the ethylene homopolymers and copolymers produced by the process and it would have been obvious for one of ordinary skill in the art to use the similar approach of Hennenberger et al. to develop another set of equations to describe the density-reactor temperature relationship for the process disclosed by Smith et al. to obtain claims 1-9.

To establish *prima facie* obviousness of a claimed invention, all the claim limitations must be taught or suggested by the prior art. *In re Royka*, 490 F.2d 981, 180 USPQ 580 (CCPA 1974). As a result of applicants' present amendments and the remarks below applicants believe the examiner has not established *prima facie* obviousness.

Applicants currently amend claim 1 to specify said metal oxide to metal oxide of

groups 1-14 of the periodic table. The support for this amendment can be found in the specification on page 6, lines 15-18.

Applicants also enclose a copy of Greenwood/Earnshaw, Chemistry of the Elements, a standard inorganic textbook. This shows that oxides are binary compounds with oxygen. Metal oxides consist of metal ions and oxide counter-ions and are thus no oxides. Phosphates are oxygen-containing compounds, but they are neither a metal oxide (they are ternary compounds) nor oxides of phosphorous but only the anion in salts of phosphorous acid. Therefore, it is evident that the aluminum phosphate of Mueller et al. is not a metal oxide.

Applicants restrict "chromium catalyst" to an "inorganic chromium catalyst and/or an organic chromium catalyst consisting of a one-core organometallic chromium compound." The support for this amendment is given on page 6, lines 29-36. The chromium compound of Smith et al. are excluded. Applicants also add a new claim containing a list of chromium compounds as listed in claim 1 of the parent patent of the present application. The support for this amendment can be found page 6, line 29-36 of the present specification.

In view of the current amendments and the foregoing, applicants believe the present claims are not obvious over Mueller et al. in view of Hennenberger et al. Otherwise, the examiner's position would be inconsistent with his previous notice of allowance wherein AlPO_4 also are excluded. Same is true for the combination of Smith et al. with Hennenberger et al. because the multi-core chromium compounds are

excluded.

For the reasons expressed above, it is urged that the prior art references cited by the examiner either singly or in combination fail to anticipate or suggest the present invention as defined by the amended claims. Accordingly, a *prima facie* case of obviousness has not been established by the examiner, and the rejection under 35 USC § 103 should be withdrawn.

Claims 5-9 are rejected under 35 USC § 102(b) as anticipated by or, in the alternative, under 35 USC § 103(a) as obvious over Mueller et al.

Applicants herein cancel claims 5-9.

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Respectfully submitted,

KEIL & WEINKAUF

A handwritten signature in black ink, appearing to read "Dan Kim", written in a cursive style.

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Chemistry of the Elements

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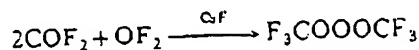
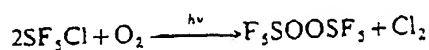
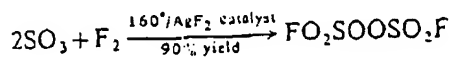
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Oxides

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the past few decades. Although such applications have not yet materialized, numerous new compounds of this type have been synthesized and characterized, e.g.:



Such compounds are volatile liquids or gases (Table 14.13) and their extensive reaction chemistry has been very fully reviewed.⁽⁹⁷⁾

TABLE 14.13 Properties of some fluorinated peroxides

Compound	MP/°C	BP/°C	Compound	MP/°C	BP/°C
FO ₂ SOOSO ₂ F	-55.4	67.1	F ₃ COONO ₂	—	0.7
FO ₂ SOOF	—	0	F ₃ COOP(O)F ₂	-88.6	15.5
FO ₂ SOOSF ₅	—	54.1	F ₃ COOCI	-132	-22
F ₃ SOOSF ₅	-95.4	49.4	(F ₃ C) ₂ COOC(CF ₃) ₂	12	98.6
F ₃ SOOCF ₃	-136	7.7	F ₃ COOOCF ₃	-138	-16

14.2.5 Oxides

Various methods of classification

Oxides are known for all elements of the periodic table except the lighter noble gases and, indeed, most elements form more than one binary compound with oxygen. Their properties span the full range of volatility from difficultly condensable gases such as CO (bp -191.5°C) to refractory oxides such as ZrO₂ (mp 3265°C, bp ~4850°C). Likewise, their electrical properties vary from being excellent insulators (e.g. MgO), through semiconductors (e.g. NiO), to good metallic conductors (e.g. ReO₃). They may be precisely stoichiometric or show stoichiometric variability over a narrow or a wide range of composition. They may be thermodynamically stable or unstable with respect to their elements, thermally stable or unstable, highly reactive to common reagents or almost completely inert even at very high temperatures. With such a vast array of compounds and such a broad spectrum of properties any classification of oxides is likely to be either too simplified to be reliable or too complicated to be useful. One classification that is both convenient and helpful at an elementary level stresses the acid-base properties of oxides; this can be complemented and supplemented by classifications which stress the structural relationships between oxides. General classifications based on redox properties or on presumed bonding models have proved to be less helpful, though they are sometimes of use when a more restricted group of compounds is being considered.

⁹⁷ R. A. DE MARCO and J. M. SHREEVE, Fluorinated peroxides, *Adv. Inorg. Chem. Radiochem.*, 16, 109-76 (1974); J. M. SHREEVE, Fluorinated peroxides, *Endeavour* xxxv, No. 125, 79-82 (1976).

The acid-base classification⁽⁹⁸⁾ turns essentially on the thermodynamic properties of hydroxides in aqueous solution, since oxides themselves are not soluble as such (p. 738). Oxides may be:

acidic: e.g. most oxides of non-metallic elements (CO_2 , NO_2 , P_4O_{10} , SO_3 , etc.);
basic: e.g. oxides of electropositive elements (Na_2O , CaO , Ti_2O_3 , La_2O_3 , etc.);
amphoteric: oxides of less electropositive elements (BeO , Al_2O_3 , Bi_2O_3 , ZnO , etc.);
neutral: oxides that do not interact with water or aqueous acids or bases (CO , NO etc.).

Periodic trends in these properties are well documented (p. 31). Thus, in a given period oxides progress from strongly basic, through weakly basic, amphoteric, and weakly acidic, to strongly acidic (e.g. Na_2O , MgO , Al_2O_3 , SiO_2 , P_4O_{10} , SO_3 , ClO_2). Acidity also increases with increasing oxidation state (e.g. $\text{MnO} < \text{Mn}_2\text{O}_3 < \text{MnO}_2 < \text{Mn}_2\text{O}_7$). A similar trend is the decrease in basicity of the lanthanide oxides with increase in atomic number from La to Lu. In the main groups, basicity of the oxides increases with increase in atomic number down a group (e.g. $\text{BeO} < \text{MgO} < \text{CaO} < \text{SrO} < \text{BaO}$), though the reverse tends to occur in the later transition element groups.

The thermodynamic and other physical properties of binary oxides (e.g. ΔH_f° , ΔG_f° , mp, etc.) show characteristic trends and variations when plotted as a function of atomic number, and the preparation of such plots using readily available compilations of data⁽⁹⁹⁾ can be a revealing and rewarding exercise.⁽¹⁰⁰⁾

Structural classifications of oxides recognize discrete molecular species and structures which are polymeric in one or more dimensions leading to chains, layers, and ultimately, to three-dimensional networks. Some typical examples are in Table 14.14; structural details

TABLE 14.14 Structure types for binary oxides in the solid state

Structure type	Examples
Molecular structures	CO , CO_2 , OsO_4 , Tc_2O_7 , Sb_2O_6 , P_4O_{10}
Chain structures	HgO , SeO_2 , CrO_3 , Sb_2O_3
Layer structures	SnO , MoO_3 , As_2O_3 , Re_2O_7
Three-dimensional structures	See text

are given elsewhere under each individual element. The type of structure adopted in any particular case depends (obviously) not only on the stoichiometry but also on the relative sizes of the atoms involved and the propensity to form π double bonds to oxygen. In structures which are conventionally described as "ionic", the 6-coordinate radius of O^{2-} (140 pm) is larger than all 6-coordinate cation radii except for Rb^+ , Cs^+ , Fr^+ , Ra^{2+} , and Tl^+

⁹⁸ C. S. G. PHILLIPS and R. J. P. WILLIAMS, *Inorganic Chemistry*, Vol. 1, Oxford University Press, Oxford, 1965; Section 14.1, Acid, base, and amphoteric character, pp. 516–23; see also ref. 95, pp. 722–29, Acid-base character of simple oxides.

⁹⁹ M. C. BALL and A. H. NORBURY, *Physical Data for Inorganic Chemists*, Longmans, London, 1974, 175 pp. G. H. AYLWARD and T. J. V. FINDLAY, *SI Chemical Data*, 2nd edn., Wiley, Sydney, 1975, 136 pp.

¹⁰⁰ R. V. PARISH, *The Metallic Elements*, Longmans, London 1977, 254 pp. (see particularly pp. 25–28, 40–44, 66–74, 128–33, 148–50, 168–77, 188–98).

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though it is approached by K^I (138 pm) and Ba^{II} (135 pm).⁽¹⁰¹⁾ Accordingly, many oxides are found to adopt structures in which there is a close-packed oxygen lattice with cations in the interstices (frequently octahedral). For "cations", which have very small effective ionic radii (say < 50 pm), particularly if they carry a high formal charge, the structure type and bonding are usually better described in covalent terms, particularly when π interactions enhance the stability of terminal M=O bonds (M = C, N, P^V, S^{VI}, etc.). Thus, for oxides of formula MO, a coordination number of 1 (molecular) is found for CO and NO though the latter tends towards a coordination number of 2 (dimers, p. 513). With the somewhat larger Be^{II} and Zn^{II} the wurtzite (4:4) structure is adopted, and monoxides of still larger divalent cations tend to adopt the sodium chloride (6:6) structure (e.g. M^{II} = Mg, Ca, Sr, Ba, Co, Ni, Cd, Eu, etc.).

A similar trend is observed for oxides of M^{IV}O₂ in Group IV of the periodic table. The small C atom, with its propensity to form p _{π} -p _{π} bonds to oxygen, adopts a linear, molecular structure O=C=O. Silicon, being somewhat larger and less prone to double bonding (p. 419), is surrounded by 4 essentially single-bonded O in most forms of SiO₂ (p. 393) and the coordination geometry is thus 4:2. Similarly, GeO₂ adopts the quartz structure; in addition a rutile form (p. 1120) is known in which the coordination is 6:3. SnO₂ and PbO₂ also have rutile structures as has TiO₂, but the largest Group IVA cations Zr and Hf adopt the fluorite (8:4) structure (p. 129) in their dioxides. Other large cations with a fluorite structure for MO₂ are Po; Ce, Pr, Tb; Th, U, Np, Pu, Am, and Cm. Conversely, the antiferrofluorite structure is found for the alkali metal monoxides M₂O (p. 97). Such simple ideas are capable of considerable further elaboration.⁽¹⁰²⁾

Nonstoichiometry

Transition elements, for which variable valency is energetically feasible, frequently show nonstoichiometric behaviour (variable composition) in their oxides, sulfides, and related binary compounds. For small deviations from stoichiometry a thermodynamic approach is instructive, but for larger deviations structural considerations supervene, and the possibility of thermodynamically unstable but kinetically isolable phases must be considered. These ideas will be expanded in the following paragraphs but more detailed treatment must be sought elsewhere.^(103, 104, 104a, 104b)

Any crystal in contact with the vapour of one of its constituents is potentially a nonstoichiometric compound since, for true thermodynamic equilibrium, the composition of the solid phase must depend on the concentration (pressure) of this constituent in the vapour phase. If the solid and vapour are in equilibrium with each other ($\Delta G = 0$) at

¹⁰¹ R. D. SHANNON, Revised effective ionic radii and systematic studies of interatomic distances in halides and chalcogenides, *Acta Cryst.* A32, 751-67 (1976).

¹⁰² A. F. WELLS, *Structural Inorganic Chemistry*, 4th edn., Oxford University Press, Oxford, 1975; Chap. 12. Binary metal oxides, pp. 439-75; Chap. 13, Complex oxides, pp. 476-515.

¹⁰³ N. N. GREENWOOD, *Ionic Crystals, Lattice Defects, and Nonstoichiometry*, Chaps. 6 and 7, pp. 111-81, Butterworths, London, 1968.

¹⁰⁴ D. J. M. BEVAN, Nonstoichiometric compounds: an introductory essay, Chap. 49 in J. C. BAILAR, H. J. EMELÉUS, R. S. NYHOLM, and A. F. TROTMAN-DICKENSON (eds.), *Comprehensive Inorganic Chemistry*, Vol. 4, pp. 453-40, Pergamon Press, Oxford, 1973.

^{104a} T. SØRENSEN, *Nonstoichiometric Oxides*, Academic Press, New York, 1981, 441 pp.

^{104b} S. TRASATTI, *Electrodes of Conductive Metallic Oxides*, Elsevier, Amsterdam, Part A, 1980, 366 pp.; Part B, 1981, 336 pp.

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